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(54) Elongated electrochemical cell combinations.

(57) A flexible, high temperature, solid oxide electrolyte electrochemical cell stack configuration is made, comprising a plurality of flattened, elongated, connected cell combinations (1), each cell combination characterized in that an interior electrode (2) having a top surface and a plurality of interior gas feed conduits (3), through its axial length, electrolyte (5) contacting the interior electrode, and exterior electrode (8) contacting electrolyte, where a major portion of the air electrode top surface (7) is covered by interconnection material (6), and where each cell has at least one axially elongated, electronically conductive, flexible, porous, metal fiber strip material (9) in electronic connection with the air electrode (2) through contact with a major portion of the interconnection material (6), the metal fiber felt being effective as a shock absorbent body between the cells.

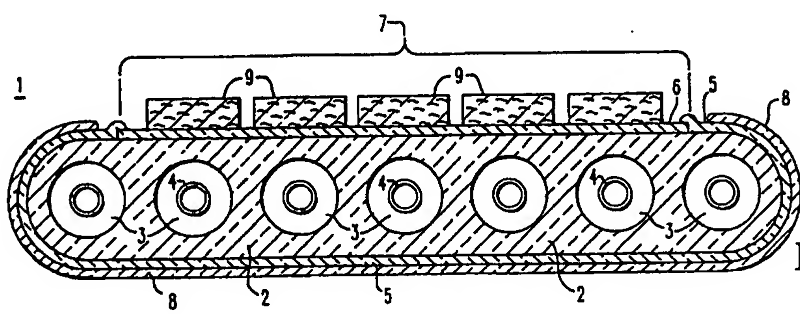

FIG. 1

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ELONGATED ELECTROCHEMICAL CELL COMBINATIONS

This invention relates to high-temperature, solid electrolyte, electrochemical cell combinations and the flexible connection of a plurality of such cell combinations.

High temperature, solid oxide electrolyte fuel cell, and fuel cell generators, are well known in the art, and are taught by Isenberg, in U.S. Patent Specification No. 4,395,468. These fuel cell configurations comprise a plurality of individual, series and parallel electronically connected, axially elongated, generally tubular, separately supported annular cells. Each cell was electronically connected in series to an adjacent cell in a column, through a narrow cell connection extending the full axial length of each cell. These connections contact the air electrode of one cell and the fuel electrode of an adjacent cell, through a conductive ceramic interconnection and a fiber metal felt strip. A single felt strip, made, for example of nickel fibers, bonded at contact points, extended axially between the cells. In the preferred embodiment air was flowed inside the cells and gaseous fuel outside.

Ackerman et al., in U.S. Patent Specification No. 4.476.198 taught a monolithic array of solid oxide electrolyte fuel cell elements. Here, triangular air and fuel conduits with surrounding electrodes and solid electrolyte were all fused together into an inflexible, ceramic matrix. A plurality of plates were stacked, with ceramic interconnects between them and the whole fused to a single rigid structure. This fused, triangular-element structure is advantageous in that it was very compact, providing a high surface area to volume area, contained no inactive materials, and did not require a separate support structure, but, it is fragile, and provides little tolerance to thermal gradients or component shrinkage during fabrication and operation. Also, a local defect caused during manufacturing or due to degradation in operation could necessitate replacement of an entire monolithic structure. The generator configuration of Ackerman et al., similarly to Isenberg, had a generating section, containing the fuel cells, disposed between an oxidant preheating section and a fuel inlet section.

None of these configurations provide a flat plate, repairable design that combines higher power density in larger individual cells, along with a flexible cell array structure that would not be sensitive to thermal gradients and stresses during start-up and operation.

It is the main object of this invention to provide a flat plate, solid oxide electrolyte electrochemical cell combination which utilizes large areas of flexible, electronically conductive, non-ceramic, metal fiber current collector materials, which would relieve thermal stress during operation of the multicell generator.

Accordingly, the invention resides in a high-temperature, solid electrolyte, flat, axially elongated electrochemical cell combination, characterized in that said cell combination comprises a flat, wide, porous, inner electrode having a top surface and a plurality of interior gas feed chambers; solid electrolyte contacting the inner electrode except for a major portion of the inner electrode top surface; outer electrode contacting the electrolyte; non-porous, ceramic, electronically conducting interconnection material, contacting the inner electrode and covering the major portion of inner electrode top surface not covered by electrolyte; and at least one axially elongated, electronically conductive, flexible, porous, metal fiber strip material in electronic connection with the inner electrode through contact with a major portion of the interconnection material.

More specifically, the cell combination comprises an air electrode having a top surface and a plurality of gas feed chambers through its cross section and parallel to its axial length, electrolyte covering the air electrode except for a major portion of the air electrode top surface, which major portion of air electrode surface is covered by non-porous, ceramic interconnection material, and a fuel electrode contacting a major portion of the electrolyte, each cell combination having at least one axially elongated electronically conductive, flexible, porous, metal fiber felt current collector material in electronic connection with the air electrode through the interconnection material.

The invention also resides in placing a plurality of these electrochemical cell combinations next to each other and, through the metal fiber felts, connecting them in series, to provide an electrochemical cell assembly. This assembly in turn can be placed in a housing where a first gaseous reactant is flowed to the air electrodes to contact the air electrodes, and a second gaseous reactant is flowed to the fuel electrodes to contact the fuel electrodes. In such an assembly, a central electrochemical cell has its fuel electrode electronically contacted in series to the air electrode of the cell below it. Said electrochemical cell has its air electrode electronically connected in series to the fuel electrode of the cell above it.

The air electrode is preferably self-supporting, and is electronically connected to the flexible, porous, metal fiber felts through an electronically conductive, non-porous, ceramic interconnection material. The cells can be of a flattened design, having circular, square, triangular, or other type geom-

etry for the interior gas conduits. This cell configuration permits large, top areas of the width of the cells to be connected, using a highly flexible, metal fiber felt, along the entire axial length of the cells, relieving stress during operation of the cell generator and making the cell stack configuration, non-rigid and non-fragile. Flattening the cell allows short electrical current paths and thinner air electrode walls, lowering gas diffusion resistance and electrical resistance. The use of large interconnection and metal fiber felt widths allows construction of more economical, larger fuel cell layers without fear of breakage due to thermal and mechanical shock. The essential, porous, metal fiber felt strip acts as a cushion as well as electronic conductor and current collector.

In order that the invention will be more readily understood, the following description of preferred embodiments will now be described, by way of example only, with reference to the accompanying drawings, in which:

Figure 1, which best illustrates the invention, is a section through a flat plate electrochemical cell combination showing a flat, extended, non-porous, conductive interconnection, and attached, extensive, flexible, porous, metal fiber top felts;

Figure 2, is a modification of the cell combination of Figure 1, showing a plurality of curved top surfaces;

Figure 3, is a section through three flat plate electrochemical cell combinations, showing flexible, porous, series connection along a major portion of each cell's width; and

Figure 4, is a section through another type of flat plate electrochemical cell combination, showing flexible, porous, series connection along the entire width of each cell.

Referring to Figure 1, a flat cross-section electrochemical cell combination 1 is shown. This flattened cell is axially elongated and contains a porous, air electrode 2, preferably self-supporting as shown, having a plurality of interior gas feed chambers 3 through its cross-section and parallel to its axial length. The air electrode top surface is shown flat in this embodiment. The gas feed chambers may, optionally, contain gas feed tubes 4, in which case the chambers 3 would be closed at one end. The gas exiting from the feed tube, into the closed end of chamber 3 would then pass through the space along the cell length to exhaust at the open end of the chamber. Preferably, the ratio of cross-sectional thickness of air electrode: cross-sectional width of the non-porous interconnection 6 shown generally as 7, of these flattened cells is from about 1:4-50. The air electrode may be a chemically modified oxide or mixture of oxides including $LaMnO_3$, $CaMnO_3$ $LaNiO_3$ and $LaCrO_3$. A preferred material is $LaMnO_3$ doped with Sr.

An interconnection 6, about 20 micrometers to about 100 micrometers thick, and typically made of lanthanum chromite doped with calcium, strontium, or magnesium, continuously covers a wide, major segment 7 along the top portion of the air electrode defining the air electrode top surface, and continues down the axial length of the air electrode. The interconnection material 6, which is a non-porous ceramic, can be as wide as the width of the air electrode, and is disposed into a discontinuity of the fuel electrode. This substantial interconnection coverage is from about 60% to about 100%, preferably about 75% to about 95%, of the air electrode cross-sectional width. The interconnection material 6 must be electrically conductive and chemically stable both in an oxygen and in a fuel environment.

The remaining balance of the porous air electrode surface is covered by a gas-tight, non-porous, solid electrolyte 5, typically yttria stabilized zirconia, about 20 micrometers to 100 micrometers thick, which is shown covering the edges of the interconnect 6 in Figure 1 to enhance gas sealing. A porous fuel electrode anode 8 contacts the electrolyte, and covers substantially the whole portion of the electrolyte. A typical anode is about 30 micrometers to 300 micrometers thick. A material (not shown) which is of the same composition as the anode, may be deposited over the interconnect 6. This material is typically nickel zirconia or cobalt zirconia cermet and is similar in thickness to that of the anode.

Figure 2 shows a modification of the cell combination of Figure 1, where the top and bottom surfaces are not flat. These surfaces can be curved as shown, or of other configuration. Such a curved surface may allow easier access of the fuel gas to the fuel electrode especially if a metal fiber mat is used for each interior gas feed chamber as shown. Figure 2 also shows the interconnection 6 covering a larger percentage of the air electrode cross-sectional width than in Figure 1.

In operation, as in the prior art, a gaseous fuel, such as hydrogen or carbon monoxide, is directed to the fuel electrode, and a source of oxygen is directed to the air electrode. The oxygen source forms oxygen ions at the electrode-electrolyte interface, which ions migrate through the electrolyte material to the anode, while electrons are supplied by the cathode, thus generating a flow of electrical current in an external load circuit. A number of cell combinations can be connected in series by contact between the non-porous interconnection 6 of one cell and the anode of another cell, through the axially elongated, electronically conductive, flexible, porous, metal fiber connection felts 9, shown covering a major portion of the interconnection ma-

terial 6.

The fibrous felt strips 9 are high-temperature stable. By "high-temperature stable" is meant that the fibrous strips contain fibers or other materials that have melting points greater than their 1000°C to 1200°C processing temperature. These strips usually have two fuel cell contacting sides which must be free of any protective coating. The strips 9 are from 80% to 97% porous (3% to 20% of theoretical density), preferably 90% to 97% porous. The felts must be electronically conducting and capable of remaining relatively flexible during fuel cell generator operation, to act as a cushion to any vibration, and to act to relieve stress and permit small displacements between the ceramic portions of the fuel cell stack during operation and cycling. The flexible, porous metal fiber connection felts are bonded fibers comprising nickel and selected from the group consisting of coated and uncoated metal fibers selected from the group consisting of nickel and cobalt fibers, preferably nickel fibers.

These fibers can range from about 0.38 cm. to 1.27 cm. long, and have a diameter of from about 0.0013 cm. to 0.025 cm. The nickel or cobalt fibers can be made by well known techniques. Final metal fiber felt strip thickness is about 0.16 cm. after compression between cells. Intermingled random orientations provide more contact between fibers and are preferred. The felt will preferably contain all nickel fibers. The body of fibers can be lightly pressed, to bring the fibers in contact with each other and then be diffusion bonded together, preferably in an inert atmosphere, such as hydrogen, argon gas. After diffusion bonding together, the bonded fibrous body can be easily handled, acquiring strength and structural integrity.

Figure 3 shows series electrical connection between adjacent fuel cell combinations that can be used in this invention. The cells 1 in the vertical column shown are electrically interconnected in series, from the inner air electrode of one cell to the outer fuel electrode of the next cell through porous metal fiber felts 9. Cumulative voltage progressively increases along the cells of a column. In Figure 3, air would be fed through the interior chambers 3 and gaseous fuel would be fed around the exterior of the cells and between the cells to contact the fuel electrodes 8. Since the fiber metal felts are from 80% to 97% porous, they can extend over a major portion, i.e., about 20% to 100% of the wide interconnection width 7, shown in Figures 1 and 2, fuel still being able to permeate the felts and contact the fuel electrodes. Figure 1 shows substantial felt coverage of the interconnection.

For the purpose of equalizing temperature and cumulative generated cell potential along the cell combination length, the longitudinal air flow direction within channels 3 may be alternated from channel to channel within each cell combination, or be uniform within each cell combination and alternated from cell combination to cell combination. Also, alternate layers of cell combinations may be translated by 90° to permit cross-flow of the air flow channels. The cell stacks would be contained within an insulation package and provided with ducting for gas supplies and exhaust, and with electrical leads for power take-off.

Figure 4 shows another variation in the electrochemical cell assembly configuration of this invention. Here, air and gaseous fuel can be fed through alternate chambers, for example, gaseous fuel can be fed through additional gas feed chambers 30 formed between the plurality of cell combinations, and air through interior chambers 31 formed by the cell stack. Here, air is kept contained by the dense electrolyte 5 and non-porous interconnection 6. Porous flexible, metal fiber felts 9 are in contact with gaseous fuel. The gaseous fuel is kept substantially isolated by the dense electrolyte 5 and non-porous interconnection 6. The wide layer of axially elongated, flexible, porous, compressible and expansible, fiber metal felt strip 9 utilized in this invention, is critical in allowing relief of thermal and mechanical stresses between ceramic portions of the cell configuration, and acts as a cushion to provide a non-monolithic structure.

Claims

1. A high-temperature, solid electrolyte, flat, axially elongated electrochemical cell combination (1), characterized in that said cell combination comprises a flat, wide, porous, inner electrode (2) having a top surface and a plurality of interior gas feed chambers; solid electrolyte (5) contacting the inner electrode except for a major portion of the inner electrode top surface; outer electrode (8) contacting the electrolyte; non-porous, ceramic, electronically conducting interconnection material (6), contacting the inner electrode and covering the major portion of inner electrode top surface not covered by electrolyte; and at least one axially elongated, electronically conductive, flexible, porous, metal fiber strip material (9) in electronic connection with the inner electrode through contact with a major portion of the interconnection material.

2. The high-temperature cell combination of claim 1, characterized in that the flexible, metal fiber strip material comprises fibers selected from the group consisting of nickel fibers and cobalt fibers.

3. The high-temperature cell combination of claim 1 characterized in that the cell combination is a fuel cell, the inner electrodes are air electrodes, the outer electrodes are fuel electrodes, the flexi-

ble, metal fiber strip material is from 80% to 97% porous, and the interconnection material coverage is from 60% to 100% of the air electrode cross-sectional width.

4. The high-temperature cell combination of claim 1, characterized in that the electrolyte is yttria stabilized zirconia, the air electrode is $LaMnO_3$, and the fuel electrode is selected from the group consisting of nickel zirconia cermet and cobalt zirconia cermet.

5. A plurality of the cell combinations of claim 1, characterized in that the inner electrode is an air electrode, the outer electrode is a fuel electrode, the interconnect on one electrode structure is electronically connected to a fuel electrode of an adjacent electrode structure, fuel is fed to contact the fuel electrodes, oxidant is fed to contact the air electrodes, and where the flexible metal fiber strip material is effective to cushion adjacent cells.

6. The plurality of cell combinations of claim 5, characterized in that the air electrode has top and bottom flat surfaces, air is fed into the interior gas feed chambers, and fuel gas is fed around the exterior of the cells to contact the fuel electrode.

7. The plurality of cell combinations of claim 5, characterized in that additional gas feed chambers are formed between the plurality of cell combinations, where air is fed into the interior gas feed chambers which contact the air electrodes and fuel gas is fed into the additional gas feed chambers which contact the fuel electrodes, and where the metal fiber strip forms a continuous shock absorbent body between the interconnection material and portions of the fuel electrode of adjacent cells.

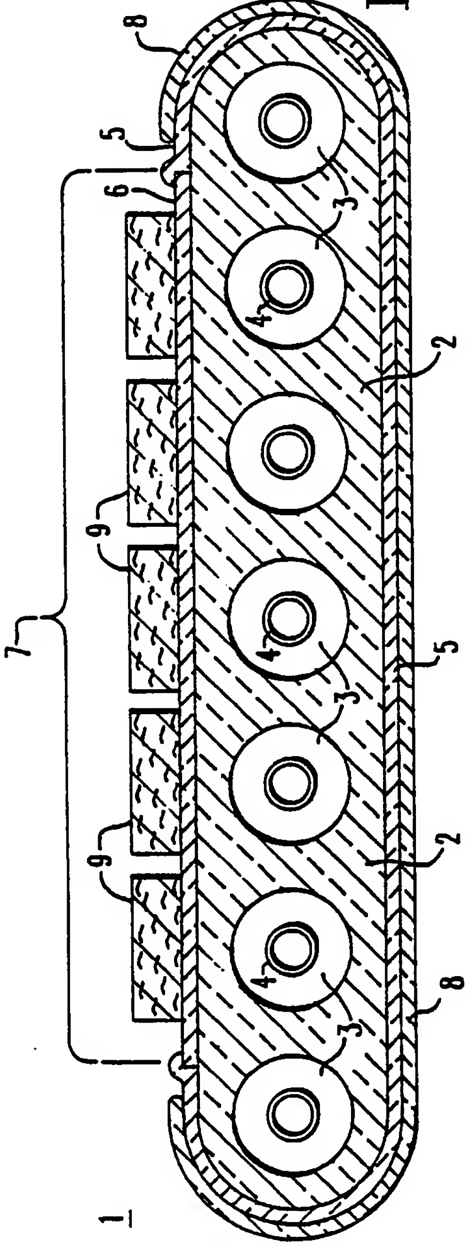

FIG. 1

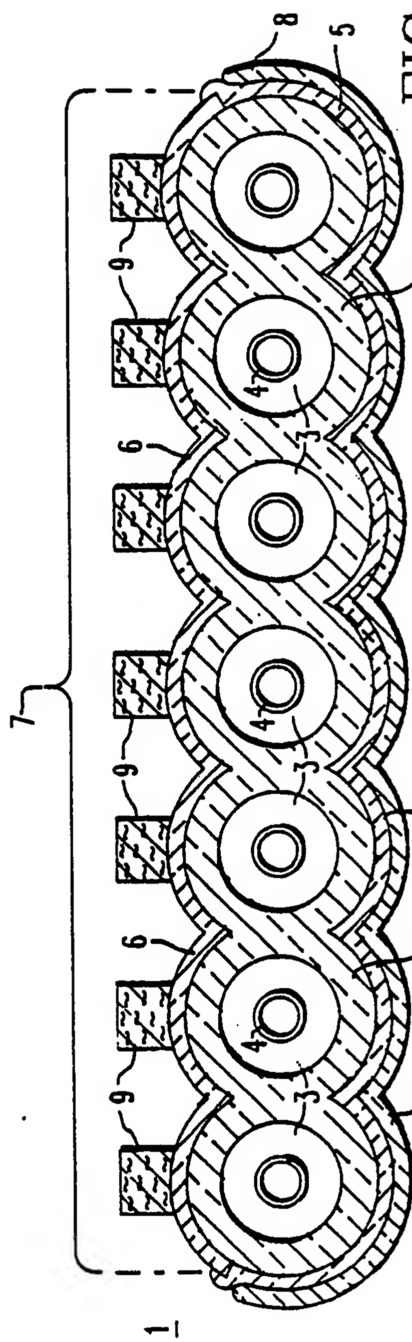

FIG. 2

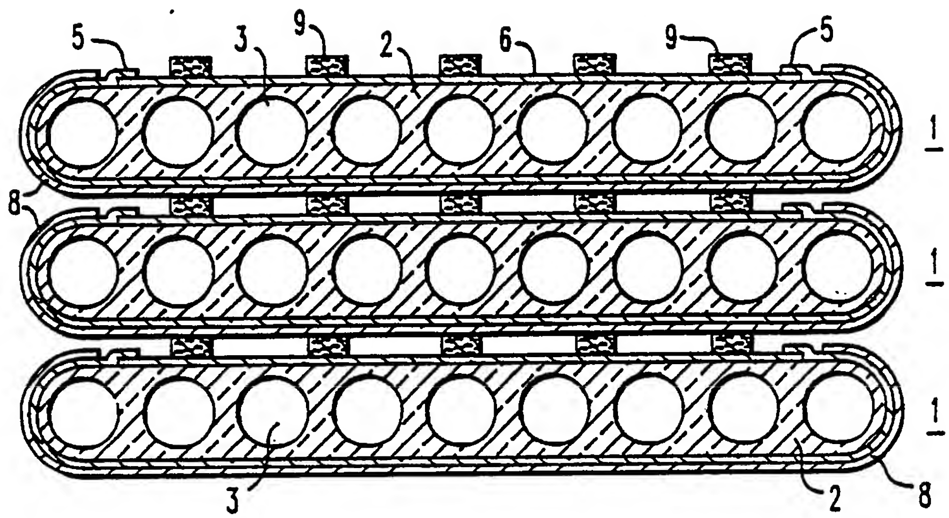

FIG. 3

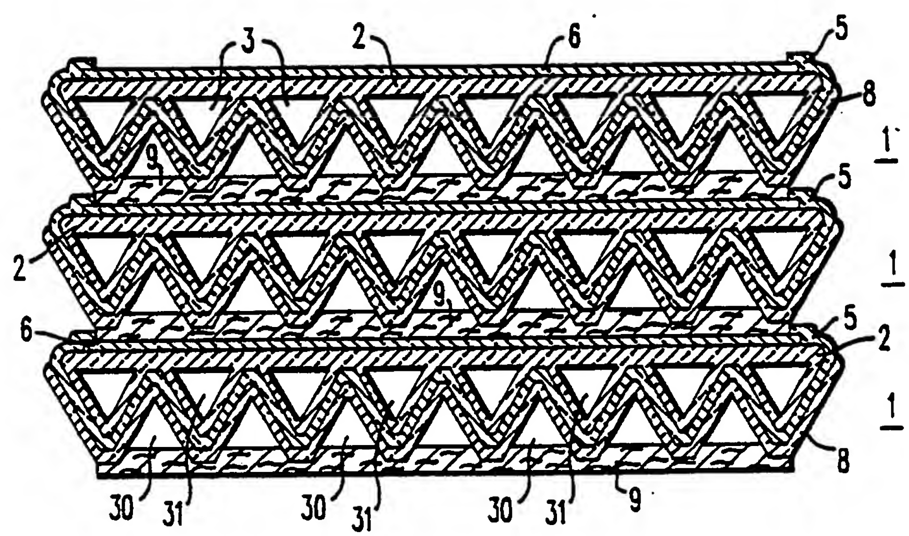

FIG. 4

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EUROPEAN SEARCH REPORT

Application Number

EP 88 30 7054

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
E,L	EP-A-0 285 727 (WESTINGHOUSE ELECTRIC CORP.) * Figures 5,6; column 6, lines 19-33; claims 1-8; figure 3; column 4, line 55 - column 5, line 1 * (Document which may throw doubt on priority claims) ---	1,2,4-6	H 01 M 8/24
X	EP-A-0 055 016 (WESTINGHOUSE ELECTRIC CORP.) * Figure 3; claims 1,3; page 7, lines 26-34; page 3, lines 12-17 * ---	1,2,4,5	
A	EP-A-0 055 011 (WESTINGHOUSE ELECTRIC CORP.) * Page 13, lines 12-14; page 7, line 29 - page 8, line 9 * & US-A-4 395 468 (Cat. A,D) ---		
P,A	US-A-4 751 152 (G.E. ZYMBOLY) * Figure 4; claims 1,6,7,10; column 5, lines 4-33 * ---		TECHNICAL FIELDS SEARCHED (Int. Cl.4)
A	GB-A-1 143 116 (BATTELLE INSTITUT) -----		H 01 M

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	31-01-1989	D'HONDT J.W.

CATEGORY OF CITED DOCUMENTS

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